

Appln. No. 10/777,229
Reply to the Office Action of November 30, 2005

Amendments to the Claims

This listing of claims will replace all prior versions, and listings, of claims in the application.

Listing of Claims

Claims 1-45. (Canceled)

Claim 46. (Currently Amended) A process for the production of aromatic hydrocarbon compounds, which comprises:

contacting one or more aliphatic hydrocarbons containing from 3 to 6 carbon atoms with a catalytic composition comprising (i) gallium, (ii) at least one lanthanide element, and (iii) a zeolite ~~belonging to~~ selected from the group consisting of the MFI[,], MEL or MFI/MEL families family of zeolites, the crystal lattice of which ~~is made up~~ consists of silicon oxide and ~~at least one metal oxide chosen from among~~ aluminum oxide in a molar ratio of silicon oxide to aluminum oxide which is greater than 20 and less than 500, boron oxide and gallium oxide and is comprised of crystallites, at least 90 % of which have diameters smaller than 500 Å.

Claim 47. (Original) A process according to Claim 46, in which the zeolite is ZSM-5.

Claims 48 and 49. (Canceled)

Claim 50. (Currently Amended) A process according to Claim 49 46, in which the ratio

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between silicon oxide and metal oxide is greater than 20 and less than or equal to 70.

Claim 51. (Currently Amended) A process according to Claim 50, in which the ratio ~~between~~ of silicon oxide to ~~and~~ metal oxide is greater than 20 and less than 60.

Claim 52. (Canceled)

Claim 53. (Previously Presented) A process according to Claim 46, in which the MFI zeolite consists of crystallites with diameters smaller than 500 Å.

Claim 54. (Previously Presented) A process according to Claim 46, in which the crystallites of the MFI zeolite present in the form of mulberry-shaped submicron aggregates with an extrazeolitic porosity of a meso-macroporous nature.

Claim 55. (Original) A process according to Claim 54, in which the total volume of said extrazeolitic porosity consists for at least 30 % of pores with diameters of less than 500 Å.

Claims 56-58. (Canceled)

Claim 59. (Currently Amended) A process according to Claim ~~58~~ 46, in which the molar ratio between silicon oxide and aluminum oxide is greater than 20 and less than or equal to 100.

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Claim 60. (Previously Presented) A process according to Claim 46, in which the zeolite is partially in acid form.

Claim 61. (Previously Presented) A process according to Claim 46, in which the catalytic composition in addition contains rhenium.

Claim 62. (Currently Amended) A process according to Claim 46 or Claim 61, in which the lanthanide is ~~chosen~~ selected from ~~among the group consisting of~~ neodymium and lanthanum, either alone or in ~~mixture~~ admixture with cerium and/or praseodymium, and mixtures of neodymium and lanthanum, optionally containing cerium and/or praseodymium.

Claim 63. (Currently Amended) A process according to Claim 46 or Claim 61, ~~in which~~ wherein the ~~hydrocarbon~~ hydrocarbon ~~said one or more~~ the aliphatic hydrocarbons containing from 3 to 6 carbon atoms are ~~chosen~~ selected from ~~among the group consisting of~~ olefins, cyclo-olefins, paraffins, and cycloparaffins.

Claim 64. (Currently Amended) A process according to Claim 46 or Claim 61, ~~in which~~ wherein the hydrocarbon or the aliphatic hydrocarbons contain from 4 to 5 carbon atoms.

Claim 65. (Currently Amended) A process according to Claim 63, in which the aliphatic hydrocarbon is ~~chosen~~ selected from ~~among the group consisting of~~ n-pentane, n-pentenes, n-butane, n-butenes, iso-butane, iso-butene, methylbutenes, cyclopentenes,

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isopentane, cyclopentane, or their mixtures.

Claim 66. (Previously Presented) A process according to Claim 46 or Claim 61, in which mixtures of aliphatic hydrocarbons are used containing from 20 to 90 wt % of olefins.

Claim 67. (Currently Amended) A process according to Claim 66, ~~in which~~ wherein mixtures of aliphatic hydrocarbons are used containing from 40 to 70 wt % of olefins.

Claim 68. (Currently Amended) A process according to Claim 46 or Claim 61, which is conducted at a temperature ranging from 300° to 800° C, and at a pressure ranging from 0 to 20 barg.

Claim 69. (Currently Amended) A process according to Claim 68, which is conducted at a temperature ranging from 400° to 650° C, and at a pressure ranging from 1 to 10 barg.

Claim 70. (Currently Amended) A process according to Claim 68, which is conducted at a WHSV ranging from 0.1 to 30 hours⁻¹.

Claim 71. (Withdrawn) A catalytic composition, comprising:

gallium,

at least one lanthanide,

and a zeolite selected from the group consisting of MFI, MEL and MFI/MEL families,

wherein the zeolite has a crystal lattice comprising silicon oxide and at least one metal

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oxide selected from the group consisting of aluminum oxide, boron oxide and gallium oxide,
and wherein the amount of the lanthanide, expressed as the element, ranges from 0.01
to 10 wt %.

Claim 72 (Withdrawn) The composition according to Claim 71, in which the zeolite is
selected from the group consisting of ZSM-5, zeolites having an MFI structure based on silicon
oxide, gallium oxide and optionally aluminum oxide, zeolites having an MFI structure based
on silicon oxide and boron oxide, ZSM-11, zeolites having a MEL structure based on silicon
oxide and boron oxide, ZSM-8, and zeolites having an MFI/MEL structure based on silicon
oxide and boron oxide.

Claim 73. (Withdrawn) The composition according to Claim 72, in which the zeolite
is ZSM-5.

Claim 74. (Withdrawn) The catalytic composition according to Claim 71, in which the
molar ratio between silica and the metal oxide is greater than 20.

Claim 75. (Withdrawn) The catalytic composition according to Claim 74, in which the
molar ratio between silicon oxide and metal oxide is greater than 20 and less than 500.

Claim 76. (Withdrawn) The catalytic composition according to Claim 75, in which the
molar ratio between silicon oxide and metal oxide is greater than 20 and less than or equal to

70.

Claim 77. (Withdrawn) The catalytic composition according to Claim 76, in which the molar ratio between silicon oxide and metal oxide is greater than 20 and less than 60.

Claim 78. (Withdrawn) The catalytic composition according to Claim 71, in which the MFI zeolite consists of crystallites with diameters smaller than 500 Å.

Claim 79. (Withdrawn) The catalytic composition according to Claim 71, in which the crystallites of the MFI zeolite are present in the form of mulberry-shaped submicron aggregates with an extrazeolitic porosity of a meso-macroporous nature.

Claim 80. (Withdrawn) The catalytic composition according to Claim 79, wherein at least 30 % of the total volume of said extrazeolitic porosity consists of pores with diameters of less than 500 Å.

Claim 81. (Withdrawn) The catalytic composition according to Claim 71, in which the crystal lattice of the MFI zeolite is made up of silicon oxide and aluminum oxide.

Claim 82. (Withdrawn) The catalytic composition according to Claim 81, in which the molar ratio molar ratio between silicon oxide and aluminum oxide is greater than 20.

Claim 83. (Withdrawn) The catalytic composition according to Claim 82, in which the molar ratio between silicon oxide and aluminum oxide is greater than 20 and less than 500.

Claim 84. (Withdrawn) The catalytic composition according to Claim 83, in which the molar ratio between silicon oxide and aluminum oxide is greater than 20 and less than or equal to 100.

Claim 85. (Withdrawn) The catalytic composition according to Claim 71, in which the zeolite is partially in acid form.

Claim 86. (Withdrawn) The catalytic composition according to Claim 71, which further comprises rhenium.

Claim 87. (Withdrawn) The catalytic composition according to Claim 71, in which the lanthanide is present in the form of an oxide, ion, metal or is a mixture of these forms.

Claim 88. (Withdrawn) The catalytic composition according to Claim 87, in which the amount of lanthanide, expressed as the element, ranges from 0.1 to 2 wt %.

Claim 89. (Withdrawn) The catalytic composition according to Claim 71, in which the lanthanide is selected from the group consisting of neodymium and lanthanum, either alone or in mixtures with cerium and/or praseodymium, and mixtures of neodymium and lanthanum,

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optionally with cerium and/or praseodymium.

Claim 90. (Withdrawn) The catalytic composition according to Claim 71, in which the gallium is present in the form of an oxide, gallium ion, metallic gallium or is a mixture of these forms.

Claim 91. (Withdrawn) The catalytic composition according to Claim 71, in which the amount of gallium, expressed as the element, ranges from 0.05 to 10 wt %.

Claim 92. (Withdrawn) The catalytic composition according to Claim 91, in which the amount of gallium, expressed as the element, ranges from 0.5 to 4 wt %.

Claim 93. (Withdrawn) The catalytic composition according to Claim 86, in which the amount of rhenium, expressed as the element, ranges from 0.05 to 10 wt % with respect to the total weight of the catalytic composition.

Claim 94. (Withdrawn) The catalytic composition according to Claim 93, in which the amount of rhenium ranges from 0.5 to 4 wt %.

Claim 95. (Withdrawn) The catalytic composition according to Claim 86, in which the rhenium is present in the form of an oxide, ion, metal or is a mixture of these forms.

Claim 96. (Withdrawn) The catalytic composition according to Claim 71, containing a binder selected from the group consisting of silica, alumina and clay, in a proportion by weight ranging from 50:50 to 95:5.

Claim 97. (Withdrawn) The catalytic composition according to Claim 71, in which the zeolite is prepared by stirring a solution containing sources of tetra-propyl-ammonium ion, sodium oxide, aluminum oxide, silicon oxide, and water, having the following composition expressed as molar ratios:

$\text{OH}^-_{\text{free}}/\text{SiO}_2$	0.07-1.0
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$(\text{C}_3\text{H}_7)_4\text{N}^+/\text{SiO}_2$	0.01-1
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$\text{H}_2\text{O}/\text{OH}^-_{\text{free}}$	10-300
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$\text{SiO}_2/\text{Al}_2\text{O}_3$	> 5
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Na^+/SiO_2	0.6-5
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up to completion of crystallization.

Claim 98. (Withdrawn) The catalytic composition according to Claim 97, in which the composition of the synthesis mixture is as follows:

$\text{OH}^-_{\text{free}}/\text{SiO}_2$	0.1-0.3
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$(\text{C}_3\text{H}_7)_4\text{N}^+/\text{SiO}_2$	0.05-0.25
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$\text{H}_2\text{O}/\text{OH}^-_{\text{free}}$	20-60
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$\text{SiO}_2/\text{Al}_2\text{O}_3$	50-120
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Na^+/SiO_2	1-4.
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Claim 99. (Withdrawn) The catalytic composition according to Claim 97, in which the crystallization is performed at a temperature in the range from 90 to 130° C, while stirring for a time which ranges from 3 hours to 15 days, and optionally comprises a second stage that can last up to 5 days in which the temperature is raised to a value ranging from 110 to 160° C.

Claim 100. (Withdrawn) The catalytic composition according to Claim 97, in which the source of the tetra- propyl-ammonium ion is selected from the group consisting of the corresponding bromide, the corresponding hydroxide, and a mixture comprising tri-n-propylamine and n-propyl bromide.

Claim 101. (Withdrawn) The composition according to Claim 97, in which the source of silicon oxide is selected from the group consisting of sodium silicate, silica hydrosol, silica gel and silicic acid.

Claim 102. (Withdrawn) The composition according to Claim 97, in which the source of aluminum oxide is selected from the group consisting of sodium aluminate, alumina, aluminum sulphate and aluminum nitrate.

Claim 103. (Withdrawn) The composition according to Claim 97, in which the source of sodium is selected from the group consisting of the corresponding hydroxide, corresponding halide, corresponding sulphate, the aluminum source, the silicon source, and mixtures thereof.

Claim 104. (Withdrawn) A process for preparing the catalytic composition according to Claim 71, which comprises treating the zeolite with a gallium compound to obtain a product, treating said product with a lanthanide compound, and then drying and calcining the product obtained.

Claim 105. (Withdrawn) A process for preparing the catalytic composition according to Claim 71, which comprises:

(1) treating the zeolite with a lanthanide compound to obtain a product, treating said product with a gallium compound, and then drying and calcining the product obtained,
or

(2) treating the zeolite with a mixture comprising a gallium compound and a lanthanide compound, and then drying and calcining the product obtained.

Claim 106. (Withdrawn) The process according to Claim 104, in which the zeolite is in acid form.

Claim 107. (Withdrawn) The process according to Claim 104, in which the treatment with a lanthanide compound is selected from the group consisting of ion exchange and impregnation.

Claim 108. (Withdrawn) The process according to Claim 107, in which the treatment with a gallium compound consists in an ion exchange or impregnation with an aqueous

solution of a gallium salt, and the treatment with a lanthanide compound consists in an impregnation with an aqueous solution of a lanthanide salt.

Claim 109. (Withdrawn) The process according to Claim 104, which comprises the treatment of the zeolite by means of an ion exchange or impregnation with an aqueous solution of a gallium salt, drying, optionally calcining the resulting product, treating by means of impregnation with an aqueous solution of a lanthanide salt and then drying and calcining the product obtained.

Claim 110. (Withdrawn) The process according to Claim 108, in which the gallium salt and the lanthanide salt are selected from the group consisting of the corresponding nitrates, chlorides and sulphates.

Claim 111. (Withdrawn) The process according to Claim 104, which comprises which comprises the treatment of the zeolite by means of ion exchange or impregnation with an aqueous solution of a gallium salt, drying the impregnated zeolite, impregnating the zeolite with an aqueous solution of a lanthanide salt and then drying and calcining the zeolite product obtained.

Claim 112. (Withdrawn) The process according to Claim 104, which comprises which comprises the treatment of the zeolite by means of ion exchange or impregnation with an aqueous solution of a gallium salt, drying the impregnated zeolite, impregnating the zeolite

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with an aqueous solution of a lanthanide salt and then drying and calcining the zeolite product obtained.

Claim 113. (Withdrawn) A process for preparing the catalytic composition according to Claim 86, which comprises treating the zeolite with a gallium compound, a lanthanide compound and a rhenium compound in any order and then drying and calcining the product obtained.

Claim 114. (Withdrawn) The process according to Claim 113, which comprises:

- a) treating the zeolite by ion exchange or impregnation with an aqueous solution of a gallium salt, and then drying and optionally calcining the impregnated zeolite;
- b) treating the impregnated zeolite by impregnation with an aqueous solution of a lanthanide salt and the drying and optionally calcining the resulting product; and
- c) treating the product by impregnation with an aqueous solution of a rhenium salt and the drying and calcining the resulting product.

Claim 115. (Withdrawn) A catalytic composition, consisting of:

gallium,
at least one lanthanide,
and a zeolite selected from the group consisting of MFI, MEL and MFI/MEL families,
wherein the zeolite has a crystal lattice comprising silicon oxide and at least one metal oxide selected from the group consisting of aluminum oxide, boron oxide and gallium oxide,

and wherein the amount of the lanthanide, expressed as element, ranges from 0.01 to 10 wt %.

Claims 116 and 117. (Canceled)

Claim 118. (New) A process according to Claim 46, wherein said lanthanide element is present in the catalyst in an amount of 0.01 to 10 wt %, based on the total weight of the catalyst composition.

Claim 119. (New) A process according to Claim 46, wherein said gallium is present in the catalyst in an amount of 0.05 to 10 wt %, based on the total weight of the catalyst composition.

Claim 120. (New) A process for the production of aromatic hydrocarbon compounds, which comprises:

contacting one or more aliphatic hydrocarbons containing from 3 to 6 carbon atoms with a catalytic composition comprising (i) gallium, (ii) at least one lanthanide element, and (iii) a zeolite selected from the group consisting of the MFI family of zeolites, the crystal lattice of which consists of silicon oxide and aluminum oxide in a molar ratio of silicon oxide to aluminum oxide which is greater than 20 and less than 500, and is comprised of crystallites, at least 90 % of which have diameters smaller than 500 Å, and wherein, in the process of introducing the zeolite with gallium and said at least one lanthanide, the process consists of

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introducing gallium and lanthanide elements into the zeolite and then drying and calcining the zeolite product obtained.

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Abstract of the Disclosure

The deletion of the existing abstract on page 73 of the disclosure is requested in favor of the new abstract presented by way of the separate sheet that shows the abstract to the present response.